

Aberystwyth University

In-situ X-ray structure measurements on aerodynamically levitated high temperature liquids

Weber, Richard; Benmore, Christopher; Mei, Qiang; Wilding, Martin

Published in:

Synchrotron Radiation in Materials Science

DOI:

[10.1063/1.3086241](https://doi.org/10.1063/1.3086241)

Publication date:

2009

Citation for published version (APA):

Weber, R., Benmore, C., Mei, Q., & Wilding, M. (2009). In-situ X-ray structure measurements on aerodynamically levitated high temperature liquids. In R. Magalhaes Paniago (Ed.), *Synchrotron Radiation in Materials Science: Proceedings of the 6th International Conference on Synchrotron Radiation in Materials Science* (Vol. 1092, pp. 79-83). (AIP Conference Proceedings; Vol. 1092). American Institute of Physics. <https://doi.org/10.1063/1.3086241>

General rights

Copyright and moral rights for the publications made accessible in the Aberystwyth Research Portal (the Institutional Repository) are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the Aberystwyth Research Portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the Aberystwyth Research Portal

Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

tel: +44 1970 62 2400

email: is@aber.ac.uk

***In-situ* X-ray structure measurements on aerodynamically levitated high temperature liquids.**

Richard Weber^{*1,2}, Christopher Benmore², Qiang Mei² and Martin Wilding³

1. Materials Development, Inc., Arlington Hts., IL 60004, USA, rweber@matsdev.com

2. Argonne National Laboratory, benmore@anl.gov, qiang.mei@hpcat.aps.anl.gov,

3. University of Wales, mbw@aber.edu

Abstract. High energy, high flux X-ray sources enable new measurements of liquid and amorphous materials in extreme conditions. Aerodynamic levitation in combination with laser beam heating can be used to access high purity and non-equilibrium liquids at temperatures up to 3000 K. In this work, a small aerodynamic levitator was integrated with high energy beamline 11 ID-C at the Advanced Photon Source. Scattered X-rays were detected with a Mar345 image plate. The experiments investigated a series of binary in the CaO-Al₂O₃, MgO-SiO₂, SiO₂-Al₂O₃ metal oxide compositions and pure SiO₂. The results show that the liquids exhibit large changes in structure when the predominant network former is diluted. Measurements on glasses with the same compositions as the liquids suggest that significant structural rearrangement consistent with a fragile-strong transition occurs in these reluctant glass forming liquids as they vitrify.

Keywords: Materials Science, Synchrotron radiation

PACS: 81.00.00

INTRODUCTION

Extreme sample environments provide unique opportunities when they are used in combination with high flux x-ray sources. The use of aerodynamic levitation with laser beam heating provides a convenient way to access very high temperature liquids in conditions that avoid contamination and heterogeneous nucleation by container walls [1,2]. This enables (i) the study of metastable high temperature liquids, and (ii) formation of new glasses. The aerodynamic levitation technique is extremely versatile and compact and it has been used to study liquid metals, oxides and integrated with NMR, neutron, and x-ray facilities [3,4].

The present research is focused on investigation of fragile binary oxide liquids. These liquids exhibit a highly non-Arrhenius temperature dependence of viscosity [5]. In many cases fragile liquids can be vitrified using levitation methods. The study of

the high temperature liquid and the corresponding glass structure provides insights in structural evolution as the liquid cools, its viscosity increases, and ultimately the glass transition occurs.

EXPERIMENTAL METHODS

A photograph of the laboratory-based aerodynamic levitation facility at the Advanced Photon Source is shown in Fig. 1. This instrument is used in the laboratory to synthesize glasses and investigate undercooling of liquids. The instrument can be installed at the high energy beamline 11 ID-C for *in-situ* measurements on liquids. The sample temperature is measured using an optical pyrometer and progress of the experiments is followed from outside the beamline hutch using video cameras that view the sample. Samples approximately 3 mm in diameter can be levitated in process gases (oxygen or argon, which is necessary for metals).

In the x-ray experiments, scattered x-rays are detected using a Mar-345 image plate that is located approximately 50 cm from the sample. The x-ray beam is 1 x 1 mm and intersects the top part of the sample in the region where it is heated and where the temperature is measured. The instrument is calibrated using a standard CeO_2 sample and then the materials to be investigated are introduced into the levitator.

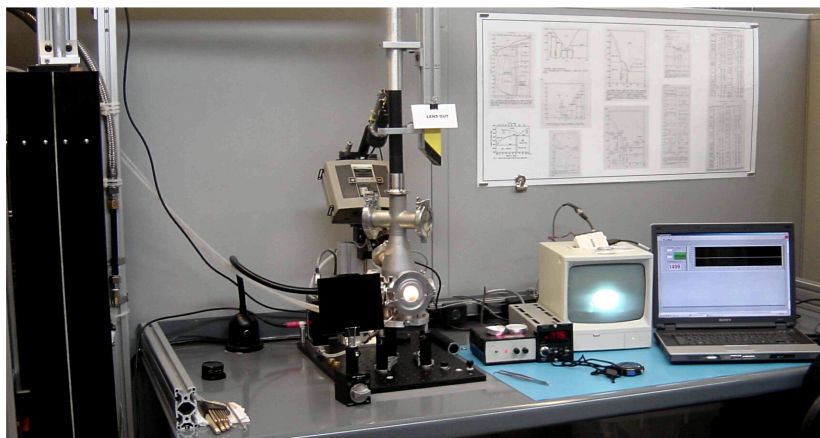


Figure 1. Image of the aerodynamic levitation facility at the Advanced Photon Source. Binary oxide compositions are made by fusing high purity metal oxide powder mixtures.

A schematic layout of the experimental set up is illustrated in Fig. 2. A tungsten pin located at the center of the image plate blocks the direct x-ray beam. Scattered x-ray data is acquired by computer and analyzed using procedures established in prior work [6].

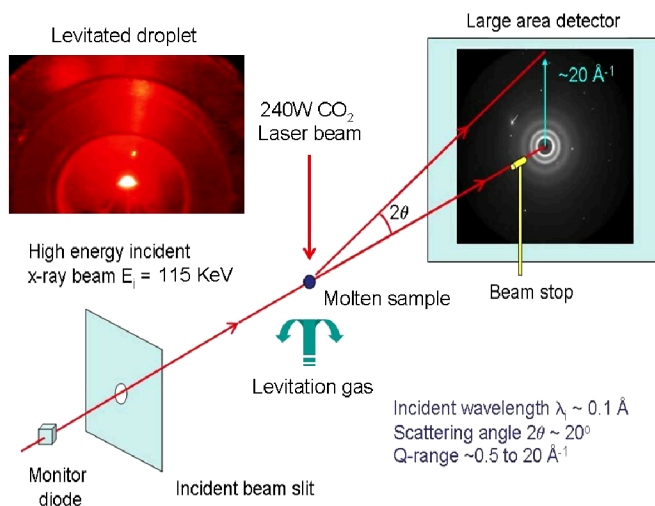


Figure 2 - Illustration of the experimental set up at the beamline. By using an incident x-ray energy of 115 keV a Q-range of *ca.* 20\AA^{-1} is obtained

Silica. High purity silica glass and the corresponding liquid were investigated at temperatures from 25-2100°C [6]. The strong liquid exhibits very minor structure changes with temperature: (i) ~2% increase in Si-O bond length from 25 to 1600°C, and (ii) the average bond angle decreases ~9° at high temperature, indicating small changes in polymer ring size

CaO-Al₂O₃ binaries. The materials melt to form fragile liquids that can be vitrified over a range from approximately 50-70 mole % CaO. The derived $D(r)$ for liquids at 2000°C and glass at ambient temperature are presented in Fig. 3.

While the Al-O first nearest neighbor distance is essentially the same in the glass and liquid, there are significant changes in the Ca-O bonding and the correlations at longer distances. NMR measurements on similar glasses indicated that AlO_4 triclusters are present in compositions containing 50% CaO but not in those with higher CaO contents [8,9]. MD simulations on the liquid [10] indicate a substantial concentration of triclusters in the liquid.

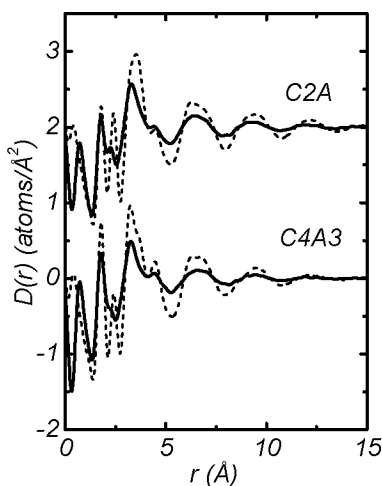


Figure 3. Comparison of the structures of liquid and glass for two compositions in the CaO-Al₂O₃ binary system shown out to long distances, glass is the dashed line – see ref [7].

MgO-SiO₂ binaries. Detailed structure measurements across the MgSiO₃-Mg₂SiO₄ region show that there is a transition from predominantly SiO₂ network to MgO network at approximately 38 mole % SiO₂ in the glass. Measurements on the corresponding high temperature liquids suggest that the transition in network behavior occurs at approximately 48 mole % SiO₂ in the liquid.

Al₂O₃-SiO₂ binaries. Measurements were made on liquids containing 30-90 mole% Al₂O₃. The melt structure is remarkably constant over this range comprising predominantly 4-coordinated Si-O and Al-O species. Simulations indicate a high population of AlO₄ triclusters in the liquids. In contrast, glasses formed in this system show high populations of 5 and 6-coordinated aluminum ions [11].

DISCUSSION

For the three binary oxide systems investigated, there are substantial differences in structure between the glass at ambient temperature and the high temperature liquid. In the case of the aluminates, the presence of triclusters of AlO₄ species appears to be common in the high temperature liquids. On cooling, the concentration of triclusters decreases and the glasses frequently contain higher coordinated aluminum species. In the magnesium silicates, changes in Mg-O bonding occur as the liquid is cooled.

The high and low temperature structural ‘snapshots’ enabled by the high energy x-ray experiments provide a new insight into the structural evolution of the liquids with temperature. Ongoing research is targeted towards understanding the temperature dependence of structure as the liquids approach the glass transition. One key goal of this work is to determine if the fragile-strong transition exhibits the character of a phase transition or a slow evolution of structure with changing temperature.

ACKNOWLEDGMENTS

This work was supported by the U.S. DOE, at Argonne National Laboratory under contract number DE-AC02-06CH11357.

REFERENCES

1. S. Ansell, S. Krishnan, J.K.R. Weber, J.J. Felten, P.C. Nordine, M.A. Beno, D.L. Price and M-L Saboungi, *Phys. Rev. Lett.*, 1997, 78, 464.
2. J. J. Wall, R. Weber, J. Kim, P. K. Liaw, and H. Choo, *Mater. Sci. Eng. A*, 2007, 445-446, 219.
3. C. Landron, L. Hennet, J.-P. Coutures, T.E. Jenkins, C. Aletru, G.N. Greaves, A.K. Soper and G. Derbyshire, *Rev. Sci. Instrum.*, 2000, 71, 1745.
4. J.K.R. Weber, C.J. Benmore, J.A. Tangeman, J. Siewenie and K.J. Hiera, *J. Neutron Res.*, 2003, 11, 113.
5. F. H. Stillinger, *Science*, 1995, 267, 1935..
6. Q. Mei, C.J. Benmore and J.K.R. Weber, *Phys. Rev. Lett*, 2007, 98, 057802.
7. Q. Mei, C.J. Benmore, J. Kim, J.K.R. Weber, J.E. Rix and M.C. Wilding, *J. Phys. Cond. Matt.*, in press.
8. D. Iuga, C. Morais, Z. Gan, D.R. Neuville, L. Cormier, and D. Massiot, *J. Am. Chem. Soc.* 2005, 127, 11540.
9. J.R. Allwardt, S.K. Lee and J.F. Stebbins, *Am. Miner.*, 2003, 88, 949.
10. S. Kohara, Spring-8, private communication.
11. S. Sen, R.E. Youngman, *J. Phys. Chem. B*, 2004 108, 7557.